

Novel Kondo-like behavior near magnetic instability in SmB_6 : temperature and pressure dependences of Sm valence

N. Emi¹, N. Kawamura², M. Mizumaki², T. Koyama¹, N. Ishimatsu³,

G. Pristáš⁴, T. Kagayama⁵, K. Shimizu⁵, Y. Osanai⁶, F. Iga^{6,7}, and T. Mito^{1*}

¹*Graduate School of Material Science, University of Hyogo, Ako, Hyogo 678-1297, Japan*

²*Japan Synchrotron Radiation Research Institute (JASRI), SPring-8, Sayo, Hyogo 679-5198, Japan*

³*Graduate School of Science, Hiroshima University, Higashi-Hiroshima, Hiroshima 739-8526, Japan*

⁴*Institute of Experimental Physics, Slovak Academy of Science, 04001 Košice, Slovakia*

⁵*KYOKUGEN, Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan*

⁶*College of Science, Ibaraki University, Mito Ibaraki 310-8512, Japan*

⁷*Graduate School of Science and Engineering, Ibaraki University, Mito, Ibaraki 310-8512, Japan*

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We report a systematic study of Sm valence in the prototypical intermediate valence compound SmB_6 . Mean Sm valence v_{Sm} was measured by the X-ray absorption spectroscopy as functions of pressure ($1 < P < 13$ GPa) and temperature ($3 < T < 300$ K). The pressure induced magnetic order (MO) of the sample was detected above $P_c = 10$ GPa by resistivity measurements. A shift toward the localized state of $4f$ electrons with increasing P and/or T is evident from an increase in v_{Sm} , analogous with several Yb heavy fermion compounds which show a pressure induced nonmagnetic-magnetic transition. However v_{Sm} is anomalously far below 3 at P_c , indicating the persistence of sizable delocalization of $4f$ electrons. Such a peculiar phenomenon has not been seen in the Yb and Ce compounds so far. In the P - T plot of a T dependent v_{Sm} component along with the MO phase, we found out Kondo like behavior that is generally observed in heavy fermions.

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The emergence of a wide variety of physical phenomena in f electron systems, namely lanthanide and actinide compounds, is still of considerable interest. One of key parameters governing the properties of the f electron systems is hybridization between conduction and f electrons (c - f hybridization). In some compounds, the effective c - f hybridization is changeable depending on external parameters, including temperature T and pressure P . The degree of localization in the f electrons results from the effective c - f hybridization, combined with other essential parameters such as the energy level of the f electrons relative to the Fermi level, which can be sensitive to the external parameters as well. In a strongly delocalized state, a nonmagnetic ground state evolves and valence fluctuations of the lanthanide and actinide ions are brought about. On the other hand, well localized f electrons tend to form a long-range magnetic order (MO) through the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction.

The measurement of the valence of lanthanide ions is one of the most informative methods to evaluate the degree of the localization: for example in the case of Ce, Sm and Yb compounds, the trivalent state of the lanthanide ions corresponds to the strong localization of the f electrons, while an increase in a divalent component (a tetravalent component for Ce) indicates the delocalization. In this letter, we report the comprehensive measurement of Sm valence in the prototypical intermediate va-

lence compound SmB_6 with average Sm valence of ~ 2.6 at room temperature and ambient pressure [1]. SmB_6 shows a semiconducting property with a narrow gap of $50 \sim 100$ K [2, 3], whose origin is probably intimately related to the effectively T dependent c - f hybridization [4]. The ground state of SmB_6 drastically changes with pressure, namely the insulating gap collapses at $P_c = 10$ GPa [5] and simultaneously an MO phase appears below ~ 12 K [6]. However the details of pressure induced changes in the f state has not been clarified despite intensive studies of this compound since 1960s.

We have estimated the mean Sm valence v_{Sm} by X-ray absorption spectroscopy (XAS) measurements for $1 < P < 13$ GPa and $3 < T < 300$ K. Valence fluctuations reflect electron dynamics in the energy scale as high as Coulomb interactions between the f electrons. Therefore, the valence measured by high-energy tools including the XAS may not be sensitive to nonmagnetic-magnetic changes in the ground state predominantly arising from low-energy magnetic correlations. However the present systematic study allows us to extract a valence component sensitive to low-energy electronic correlations which seems to control the ground state in this compound. We also found out the peculiarity in SmB_6 that sizable delocalization of $4f$ electrons persist at P_c , which is distinct from conventional nonmagnetic-magnetic transitions observed in Ce and Yb compounds.

Single crystalline samples of SmB_6 were grown by a floating-zone method using an image furnace with four xenon lamps [7]. The XAS measurements near the Sm L_3 -edge (6.72 keV) were performed at the beam-

*Electronic address: mito@sci.u-hyogo.ac.jp

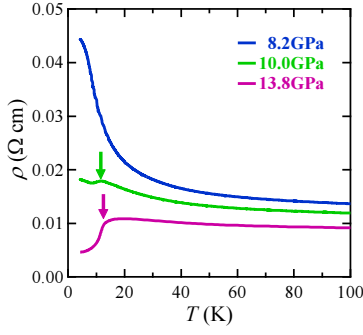


FIG. 1: (color on line) T dependence of the resistivity ρ at 8.2, 10.0, and 13.8 GPa. The arrow indicates $T_M \sim 12$ K. Here, T_M is determined as the temperature at which $d^2\rho/dT^2$ shows a minimum. For details, see Ref. [10].

line BL39XU of SPring-8, Japan [8]. Since the XAS spectrum was slightly dependent on the thickness of sample, we used a piece of SmB_6 crystal having partly constant thickness less than $10 \mu\text{m}$. The XAS spectra were recorded in the transmission mode using ionization chambers. For the high pressure measurements, the sample was loaded in a diamond anvil cell (DAC) filled with a mixture of 4 : 1 methanol : ethanol as a pressure-transmitting medium. Nanopolycrystalline diamond anvils were used to avoid glitches in the XAS spectra [9]. All pressures were applied at room temperature, and the data were acquired as the pressure cell was heated from the lowest temperature ~ 3 K. The calibration of pressure was performed at each temperature of the measurement using the fluorescence from ruby chips mounted with the sample inside the DAC. We have also carried out the high pressure measurement of the resistivity in order to detect the MO above P_c as done in Ref. [5]. For the resistivity measurement, we used a single crystalline sample from the same batch with that for the XAS measurement, and the measurement was performed using the D.C. four-terminal method. The high pressure was generated using a DAC with NaCl as the pressure-transmitting medium, and the pressure was calibrated at the lowest temperature of each measurement by the ruby fluorescence method.

Figure 1 shows the T dependence of the electrical resistivity ρ measured at different pressures around P_c . For $P < 10$ GPa, $\rho(T)$ reveals a semiconducting increase upon cooling down to ~ 7 K, followed by a tendency of saturation at lower temperatures. Although we cannot discuss the absolute value of ρ in detail due to ambiguity in the measurement of sample size, it is obvious that the semiconducting behavior is weakened near P_c and $\rho(T)$ above 10.0 GPa exhibits a drop at ~ 12 K [10]. This phenomenon and the boundary temperature $T_M \sim 12$ K and pressure $P_c \sim 10$ GPa are in good agreement with the previous report [5], indicating the appearance of the MO above P_c in our sample as well.

Figure 2(a) shows the Sm L_3 -edge absorption spectra of SmB_6 . The intensity of the main peak at 6.72

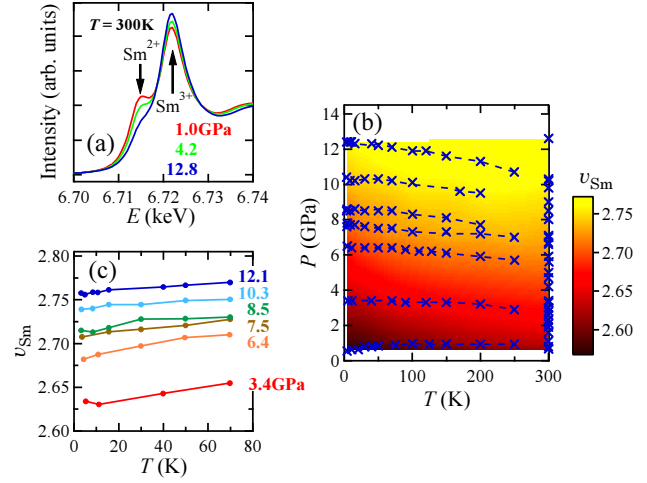


FIG. 2: (color on line) (a) Sm L_3 -edge absorption spectra of SmB_6 at 300 K and various pressures. (b) P and T dependences of estimated Sm valence v_{Sm} . The series of crosses connected by the broken lines are data points actually measured. (c) T dependence of v_{Sm} below 70 K at different pressures from 3.4 to 12.1 GPa. The clamped pressures are regarded as being almost T independent in this low temperature region.

keV, corresponding to a trivalent component, tends to increase with increasing pressure. Simultaneously, the shoulder like structure at 6.715 keV, corresponding to a divalent component, is gradually suppressed. A similar change in the XAS spectrum is also observed when temperature is increased at low constant pressures. This T variation is considerably weakened under high pressure (see Ref.[10] for details). The observation of the two components is attributed not to inhomogeneous distribution of the two valence states but to valence fluctuation at a time scale slower than that probed by XAS [1]. We further discuss the high pressure XAS data on the basis of this interpretation. v_{Sm} is estimated from the relative intensities of the Sm^{2+} and Sm^{3+} components in the XAS spectra. Each component was modeled by the sum of Lorentz functions and an arctangent function representing the continuum excitations. This commonly used method for analyzing XAS data allows a more reliable comparison between valences estimated from the present and previous works.

The evaluated v_{Sm} is illustrated in Fig. 2(b) as functions of pressure and temperature. The data points actually measured are also marked with the crosses. The clamped pressure at room temperature inevitably varies with temperature (in most cases, the pressure decreases upon heating). Therefore, we assumed a linear relation in between the neighboring v_{Sm} data points to construct this contour plot. For $T < 70$ K, we can regard the pressures to be almost constant as shown in Fig. 2(b). v_{Sm} estimated in this T -range is presented in Fig. 2(c).

We extracted vertical $v_{\text{Sm}}-T$ planes at constant pressure from Fig. 2(a), which is presented in Fig. 3(a). As temperature increases at 1 GPa, v_{Sm} increases with a

slight downward curvature, and the $v_{\text{Sm}}-T$ curves are more monotonic and smoother over the whole T range than shown in a previous report [11]. With increasing pressure, v_{Sm} in the relatively higher temperature region (*e.g.* $T > 150$ K at 6 GPa) becomes less T dependent, while v_{Sm} at low temperatures (*e.g.* $T < 50$ K at 6 GPa) is still clearly dependent on temperature. The T dependent part is gradually suppressed with pressure. Such a pressure induced change in v_{Sm} is clearly demonstrated in Fig. 3(b), where we plot $\delta v_{\text{Sm}}(T) = v_{\text{Sm}}(T) - v_{\text{Sm}}(300\text{K})$ at various pressures. Although $\delta v_{\text{Sm}}(T)$ at 12 GPa still shows a small decrease below ~ 50 K, $|\delta v_{\text{Sm}}(4\text{K})|$ is reduced by about 70 % compared to that at 1 GPa. Therefore one expects that $|\delta v_{\text{Sm}}(4\text{K})|$ will be further suppressed at higher pressures than 12 GPa. In Fig. 3(c), we plot $-\delta v_{\text{Sm}}(4\text{K})$ as a function of pressure. $\delta v_{\text{Sm}}(4\text{K})$ is extrapolated to zero at $P^* \sim 15$ GPa, indicating that v_{Sm} hardly depends on temperature above P^* .

The P dependence of v_{Sm} at constant temperature is shown in Fig. 3(d). As pressure increases, v_{Sm} increases but the slope dv_{Sm}/dP gradually decreases. Similar phenomenon is also observed in several Yb heavy fermions which show the pressure induced nonmagnetic-magnetic transition (see Ref. [10] or for example Refs. [12–14]). In the case of the Yb compounds, the saturating tendency of the Yb valence, v_{Yb} , above critical pressure is understandable, because their v_{Yb} is mostly driven by pressure to $v_{\text{Yb}} > 2.9$, close to the upper end of the valence region that Yb ions can stably take (*i.e.* $2 \leq v_{\text{Yb}} \leq 3$). Here, Yb divalent (trivalent) state is nonmagnetic (magnetic). In YbRh₂Si₂, which shows an antiferromagnetic order at extremely low temperature of 70 mK and hence is thought to locate in the vicinity of nonmagnetic-magnetic criticality at ambient pressure [15, 16], v_{Yb} is larger than 2.9 as well [17]. On the other hand, this understanding is not applicable to the present case of SmB₆, since v_{Sm} at P_c is anomalously lower than 3 ($v_{\text{Sm}} < 2.8$).

The proximity of v_{Yb} to 3 in these Yb compounds can be brought about by the strong localization of $4f$ electrons due to the marked lanthanide contraction characteristic of the Yb systems. For Ce systems, Ce valence, v_{Ce} , tends to deviate from 3 ($4f^1$) toward 4 ($4f^0$) with pressure and the lanthanide contraction should be smaller than in the Yb systems. CePd₂Si₂ and CeCu₂Ge₂ are driven through the magnetic-superconducting transition by pressure, and their v_{Ce} remains from 3.0 to 3.05 over the superconducting region (see Ref. [10] or Refs. [18, 19]). Although, for the Ce compounds, there are only a few reports of the valence measurement under pressure so far, we have found no serious exception in the Ce and Yb compounds in terms of the proximity to the trivalent state at the magnetic instability.

Such a large deviation of v_{Sm} from 3 near the magnetic instability is also observed in other Sm compounds. SmS exhibits the pressure induced MO from the intermediate valence state (the so called “gold phase”) at $P_c \sim 2$ GPa [20], and v_{Sm} at P_c is comparable to that of SmB₆ [21, 22]. SmOs₄Sb₁₂, which is the weak ferromag-

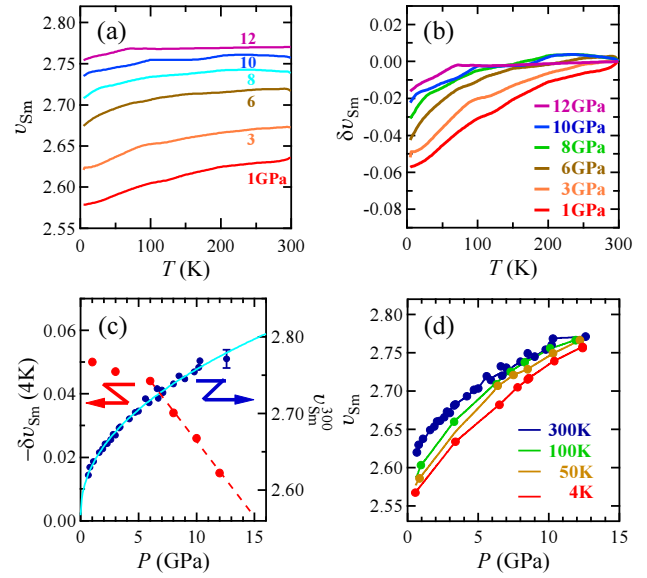


FIG. 3: (color on line) (a) $v_{\text{Sm}}-T$ curves at representative constant pressures obtained from Fig. 2(b). (b) T dependence of $\delta v_{\text{Sm}}(T) = v_{\text{Sm}}(T) - v_{\text{Sm}}(300\text{K})$ evaluated from Fig. 3(a). (c) The plots of $\delta v_{\text{Sm}}(4\text{K})$ (left axis) and $v_{\text{Sm}}^{300}(P)$ (right axis) as a function of pressure. The broken line is a linear fit to the data for $P \geq 6$ GPa. The solid line represents a fit $v_{\text{Sm}}^{300}(P) = AP^\alpha + v_{\text{Sm}}^{300}(0)$ to the data, giving $\alpha = 0.47$ and $v_{\text{Sm}}^{300}(0) = 2.57$. (d) P dependence of v_{Sm} at 4, 50, 100, and 300 K.

net with $T_C = 3$ K, also shows $v_{\text{Sm}} \sim 2.8$ [23]. Note that Sm ion possesses much smaller effective moment ($0.845\mu_B$) in the magnetic trivalent state than those of the Ce^{3+} and Yb^{3+} ions (2.54 and $4.54\mu_B$, respectively). Moreover, in these Sm compounds, the effective moment should be reduced due to the intermediate valence character. Nevertheless, magnetic ordering temperatures of SmB₆ and SmS are as high as $10 \sim 20$ K (Ref. [24]. Also see Ref. [25]). Interestingly, some of Eu compounds also show the MO away from the magnetic divalent state: for instance $\text{EuCu}_2(\text{Si}_x\text{Ge}_{1-x})_2$ with $0 < x < 0.65$ shows an antiferromagnetic order at Eu valence $2.1 < v_{\text{Eu}} < 2.4$ [26]. Therefore one may speculate that $4f^6$ state common to both Sm and Eu ions plays any role in assisting the long-range MO. However considering contribution of its excited magnetic state (total angular momentum $J = 1$) seems unrealistic, because it should be extremely small due to a Boltzman factor involving excitation energy Δ (for instance, $\Delta = 420$ K between the energy levels of $J = 0$ and 1 for the divalent Sm ion [27]).

According to Fig. 3(a), the T dependence of v_{Sm} is characterized by two features: (i) an almost T independent term seen in the higher temperature and higher pressure region, and (ii) a T dependent part in the rest region. For the latter, the $v_{\text{Sm}}-T$ curve depends on pressure as well, as indicated by Fig. 3(b). $v_{\text{Sm}}(P, T)$ is therefore described as the sum of the T and P dependent term $\Delta v_{\text{Sm}}(P, T)$ and the T independent term $v_{\text{Sm}}^*(P)$;

$$v_{\text{Sm}}(P, T) = \Delta v_{\text{Sm}}(P, T) + v_{\text{Sm}}^*(P). \quad (1)$$

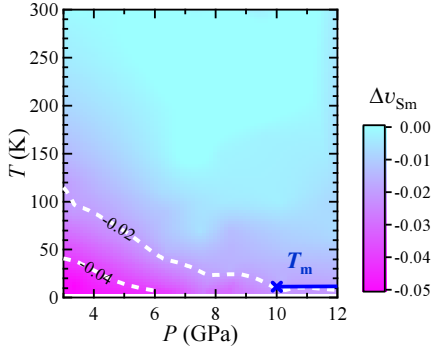


FIG. 4: (color on line) P (horizontal axis) and T (vertical axis) dependences of Δv_{Sm} . The cross and solid line indicate T_M obtained from the present resistivity measurement.

Since $|\Delta v_{\text{Sm}}(P, T)|$ is easily suppressed by the thermal effects of a few hundred Kelvins, the first term should be connected with the evolution of low-energy electronic correlations that can lead to the modification of the Sm valence, including Kondo effect. On the other hand, $v_{\text{Sm}}^*(P)$ is plausibly P dependent even for $P > P^*$ as described later, suggesting that $v_{\text{Sm}}^*(P)$ is more directly related to the valence fluctuations in higher-energy scheme through the large c - f hybridization. The existence of the large hybridization may be consistent with predictions by the band structure calculations [4, 28]. Note that, in the above-mentioned Yb and Ce compounds, the T independent term, corresponding to $v_{\text{Sm}}^*(P)$ in the case of SmB_6 , may be less P dependent and close to 3, implying that the valence fluctuations are considerably suppressed.

In order to evaluate $\Delta v_{\text{Sm}}(P, T)$, we use the relation $v_{\text{Sm}}^*(P) \cong v_{\text{Sm}}^{300}(P)$ for $P > 3$ GPa, where $v_{\text{Sm}}^{300}(P)$ is the data at 300 K. $v_{\text{Sm}}^{300}(P)$ is consistent with a recent measurement of the resonant x-ray emission spectroscopy [29]. Since $v_{\text{Sm}}^{300}(P)$ approximately follows a function of P^α with $\alpha \sim 0.5$ [10], we fit a power-law form $v_{\text{Sm}}^{300}(P) = AP^\alpha + v_{\text{Sm}}^{300}(0)$ to the data, giving $\alpha = 0.47$ and $v_{\text{Sm}}^{300}(0) = 2.57$ (the solid line in Fig. 3(c) (right axis)). $v_{\text{Sm}}^*(P)$ is unlikely to saturate even at P^* . Then $\Delta v_{\text{Sm}}(P, T)$ is extracted using Eq. (1), which is equivalent to δv_{Sm} shown in Fig. 3(b). Obtained $\Delta v_{\text{Sm}}(P, T)$ is shown in Fig. 4, where we plot pressure and temperature in the horizontal and vertical axes, respectively. Apparently the low temperature state possessing finite $\Delta v_{\text{Sm}}(P, T)$ is largely suppressed by pressure of $7 \sim 10$ GPa, in good agreement with P_c [34]. Figure 4 suggests that, if the electronic system in SmB_6 is cooled with little $\Delta v_{\text{Sm}}(P, T)$, which is the case for $P \geq 10$ GPa, it falls into the magnetically ordered ground state. Therefore the evolution of low-energy electronic correlations seems to impede the long-range MO. The emergence of the MO before $\Delta v_{\text{Sm}}(P, T)$ vanishes down to the lowest temperature may reflect the first-order nature at P_c [5, 6]. Interestingly, $v_{\text{Sm}}^*(P)$ gives a dominant contribution to the P and T dependences of $v_{\text{Sm}}(P, T)$ (i.e. $3 - v_{\text{Sm}}^* > 0.2 \gg |\Delta v_{\text{Sm}}|$), the term of $\Delta v_{\text{Sm}}(P, T)$ seem-

ingly plays a key role in determining the ground state in this compound. Such a regime well accounts for the fact that the onset of the MO does not involve any significant anomaly in v_{Sm} for $P > P_c$ where $\Delta v_{\text{Sm}}(P, T)$ is significantly suppressed (see Fig. 2(c)). Namely the MO at low temperatures should be more directly related to the low-energy electronic correlations.

Figure 4 resembles the general phase diagram expected for the heavy fermions where the so called Kondo lattice state is replaced with an MO phase as the localization of f electrons is increased by external parameters such as pressure. However one of the unique properties in SmB_6 is that $v_{\text{Sm}}^*(P)$ sizably deviates from 3 at the magnetic instability where Δv_{Sm} is fairly suppressed by pressure. This may be the issue of whether the MO by the seemingly delocalized f electrons can be accounted for in the framework of the conventional RKKY interactions. One may also need to consider a peculiar mechanism such as localized and delocalized f electrons coexist in multi f electrons system, as proposed in uranium compounds [35]. Recently, Barla *et al.* [6] suggested that, in the low pressure region, the Kondo energy $k_B T_K$ prevails over crystalline electric field splitting [36] and magnetic moments cannot interact due to their uncorrelated fast fluctuations. In this context, the extracted suppression of Δv_{Sm} at high pressures in Fig. 4 should be favorable for the appearance of long-range MO, because it is plausibly related to the decrease in T_K . In order to uncover the mechanism of the MO, the measurement of the magnitude of ordered moments will be indispensable.

In summary, we have carried out XAS measurements in the ranges of $1 < P < 13$ GPa and $3 < T < 300$ K to estimate the Sm valence of SmB_6 . Moreover the pressure induced MO of the sample was detected for $P > P_c = 10$ GPa by resistivity measurements. v_{Sm} increases with pressure and/or temperature, indicating the localization of $4f$ electrons. Although this trend is analogous with several Yb heavy fermion compounds which show a pressure induced nonmagnetic-magnetic transition, $2.7 < v_{\text{Sm}} < 2.8$ at the magnetic-nonmagnetic phase boundary is remarkably different from the known cases in the Yb and Ce compounds. The decomposed T dependent part of v_{Sm} , which is related to the evolution of low-energy electronic correlations, including Kondo effect, predominantly controls the ground state.

Acknowledgments

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- [1] The intermediate valence state in this compound is generally interpreted as arising from valence fluctuations. In the x-ray absorption spectroscopy, two sets of lines corresponding to Sm^{2+} and Sm^{3+} states are present [30]. On the other hand, in ^{149}Sm Mössbauer measurement, only one Mössbauer line is observed at an intermediated position between isomer shifts expected for the Sm^{2+} and Sm^{3+} [31]. These results indicate that the Sm valence fluctuates at the time scale in between those probed by the Mössbauer and by the x-ray absorption experiments. For a review, see for example Ref. [32].
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- [36] The resolution of the present XAS measurement is about $0.1 \sim 1$ eV, which is much larger than the realistic crystalline electric field (CEF) excitations of the order of $1 \sim 10$ meV. Therefore, even if effective CEF exists, we always count f electrons in all the CEF-split levels, leading to a conclusion that the observed large deviation of v_{Sm} from 3 at P_c is not ascribed to the CEF effect.